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An assessment of temporal trends in mercury concentrations in fish

Thomas M. Grieb[1] · Nicholas S. Fisher[2] · Roxanne Karimi[2] · Leonard Levin[3]

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Abstract

The importance of fish consumption as the primary pathway of human exposure to mercury and the establishment of fish consumption advisories to protect human health have led to large fish tissue monitoring programs worldwide. Data on fish tissue mercury concentrations collected by state, tribal, and provincial governments via contaminant monitoring programs have been compiled into large data bases by the U.S. Environmental Protection Agency's Great Lakes National Monitoring Program Office (GLNPO), the Ontario Ministry of the Environment's Fish Contaminants Monitoring and Surveillance Program (FMSP), and many others. These data have been used by a wide range of governmental and academic investigators worldwide to examine long-term and recent trends in fish tissue mercury concentrations. The largest component of the trend literature is for North American freshwater species important in recreational fisheries. This review of temporal trends in fish tissue mercury concentrations focused on published results from freshwater fisheries of North America as well as marine fisheries worldwide. Trends in fish tissue mercury concentrations in North American lakes with marked overall decreases were reported over the period 1972–2016. These trends are consistent with reported mercury emission declines as well as trends in wet deposition across the U.S. and Canada. More recently, a leveling-off in the rate of decreases or increases in fish tissue mercury concentrations has been reported. Increased emissions of mercury from global sources beginning between 1990 and 1995, despite a decrease in North American emissions, have been advanced as an explanation for the observed changes in fish tissue trends. In addition to increased atmospheric deposition, the other factors identified to explain the observed mercury increases in the affected fish species include a systematic shift in the food-web structure with the introduction of non-native species, creating a new or expanding role for sediments as a net source for mercury. The influences of climate change have also been identified as contributing factors, including considerations such as increases in temperature (resulting in metabolic changes and higher uptake rates of methylmercury), increased rainfall intensity and runoff (hydrologic export of organic matter carrying Hg^{II} from watersheds to surface water), and water level fluctuations that alter either the methylation of mercury or the mobilization of monomethylmercury. The primary source of mercury exposure in the human diet in North America is from the commercial fish and seafood market which is dominated (>90%) by marine species. However, very little information is available on mercury trends in marine fisheries. Most of the data used in the published marine trend studies are assembled from earlier reports. The data collection efforts are generally intermittent, and the spatial and fish-size distribution of the target species vary widely. As a result, convincing evidence for the existence of fish tissue mercury trends in marine fish is generally lacking. However, there is some evidence from sampling of large, long-lived commercially-important fish showing both lower mercury concentrations in the North Atlantic in response to reduced anthropogenic mercury emission rates in North America and increases in fish tissue mercury concentrations over time in the North Pacific in response to increased mercury loading.

Keywords Mercury · Fish · Temporal trends · Monitoring

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✉ Thomas M. Grieb
tom.grieb@tetratech.com

[1] Tetra Tech, Inc., Lafayette, CA, USA

[2] School of Marine and Atmospheric Sciences, State University of New York, Stony Brook, NY, USA

[3] Electric Power Research Institute, Palo Alto, CA, USA

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Introduction

There has been a major effort since the 1970s to monitor mercury concentrations in fish tissue by federal, state, provincial, and local agencies in the U.S. and Canada, with the purpose of identifying geographic areas with fish mercury concentrations at levels of potential concern for human health. More recently, numerous data sets, based on these monitoring programs, have been compiled and used to evaluate trends in mercury in fish from freshwater and marine ecosystems. In many cases, the fish mercury data have been compared to mercury deposition data from the Mercury Deposition Network (MDN) and mercury emissions data to assess the effects of local regulatory controls of mercury releases to the atmosphere as well as the influence of global atmospheric mercury emissions.

Since 1990, mercury emissions from Europe and North America have decreased while total global anthropogenic mercury emissions have increased. For example, anthropogenic mercury emissions in the U.S. (Fig. 1) have declined from 223 Mg (10^6 g) in 1990 to 92 Mg in 2005 to 51 Mg in 2011 (Weiss-Penzias et al. 2016; US EPA 2015). During the period 1990–1995 global anthropogenic emissions increased an estimated 17 to 19%, largely due to the increase of mercury emissions from Asia (Pacyna et al. 2006; Streets et al. 2009; Pirrone et al. 2010). Mercury concentrations in the North Pacific intermediate waters have increased at a rate of 3%/year between 1995 and 2006 (Drevnick and Brooks 2017). In the North Atlantic, mercury concentrations peaked in the 1980s or 1990s and were declining between 1980 and 2010 (Driscoll et al. 2013).

This study examined global temporal trends in fish tissue mercury concentrations in freshwater and marine systems based on a review of the published literature between 1970 and 2017. The relationships between trends in fish tissue mercury concentrations and mercury emissions were also examined.

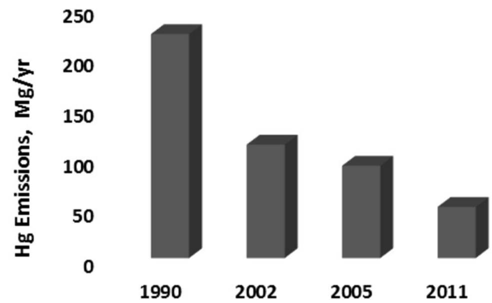

Fig. 1 U.S. mercury point source emissions (Mg/yr) 1990–2011 (Weiss-Penzias et al. 2016; US EPA 2015)

Methods

The literature search was conducted in a series of steps. The first step was conducted using the *Google Scholar* database for the period 1970–2016. The search terms initially used were: “mercury”, “temporal trend”, AND “fish” (2,610 references identified) and “mercury”, “temporal trend”, AND “fish tissue” (315 peer-reviewed journal articles, government reports and theses). Next, the results from these primary searches were reviewed, and, from the initial searches and culling of the abstracts, the most relevant published documents were identified. In the second step, this initial list was expanded by including additional references from the Seafood Mercury Database (Karimi et al. 2012). In the third step, the references within each relevant document as well as references citing each individual document (identified using Google Scholar “Cited by” option) were used to identify and obtain additional references. In the last step, the top three journals, based on the number of identified references, were searched through *Science Direct* and other search engines using title, abstract, and keyword (TITLE-ABSTR-KEY) searches for “fish”, “mercury”, and “trend” for the same time period. The literature search was conducted in October 2016, although the database includes several papers from 2017 that were identified after the initial review.

In many cases the results of the trend studies in our review included the reporting of the annual percent change (APC) in average fish tissue concentrations. In other cases, we used the average fish tissue concentration data gathered directly from tables or by digitizing plotted data presented in the reports or the supplemental data to calculate the APC. The APC was calculated using the equation $(e^B - 1) \times 100$, where B is the slope of a linear trend in log-transformed data (Hirsch et al. 1991).

Our analyses included the evaluation of trends in mercury deposition data obtained from the Mercury Deposition Network (MDN), a component of the U.S. National Atmospheric Deposition Program (NADP 2019). Trends were evaluated using the non-parametric Seasonal Mann-Kendall (SMK) test (Gilbert 1987). Monthly sums of weekly deposition measurements were used as input to the SMK. As part of the test results verification effort, the trend analyses were conducted using both the RKT package in R (R Development Core Team 2008) and the methodology described in the US EPA Unified Guidance (US EPA 2009). In the Unified Guidance the mathematical procedure described in the section titled “Identifying trends amidst seasonality: Seasonal Mann-Kendall” on pages 14–37 to 14–40 was applied.

Results and discussion

Literature summary

More than 200 references were initially identified and reviewed, and 156 references were included in the final analyses. A list of these references is presented in Table S-1. Year of publication ranged from 1972 to 2017 with increasing numbers of papers beginning in 2004 and with the greatest numbers of papers published in 2007 and 2010 (Fig. 2). There were 56 publications represented in the literature review, including 43 peer-reviewed journals represented by between one and 24 papers each. Publications also included books (n = 1), thesis manuscripts (n = 3), and various agency reports (n = 14), including reports for the U.S. Geological Survey, National Oceanic and Atmospheric Administration (NOAA), and the U.S. Environmental Protection Agency (US EPA).

Summary of reported trends

The metadata from the review of individual references were captured in a summary table. The categories of recorded data are presented and described in Table S-2. The data categories in the table were created to facilitate the compilation of data that could be used to characterize the extent and nature of the information from the literature and to quantify the trends in the fish tissue mercury concentration data. As described below, more detailed versions of the overall summary table were created for the two largest subsets of the identified papers: North American freshwater fish and marine fish. The data from these two subsets were sufficient to support detailed examination of the reported trends. The references in these two subsets and the available supplementary data were reviewed in greater detail, and the information in each of the categories described in Table S-2 was expanded.

Assessment of trends in North American freshwater fish

Approximately 25% of the identified published studies focused on trends in North American freshwater fish. The record of fish tissue mercury measurements covers the period 1967–2015. Figure 3 shows the period covered by the individual publications, ranked by duration of the study. Generally, the individual trend reports cover an extensive period. The longest record in the database is 45 years. The average years of data in the reviewed reports was 24 years, with 50 percent of the values between 14 and 35 years. The reports include some site-specific studies characterized by smaller data sets, and some regional studies characterized by very large data sets and the combining of data from

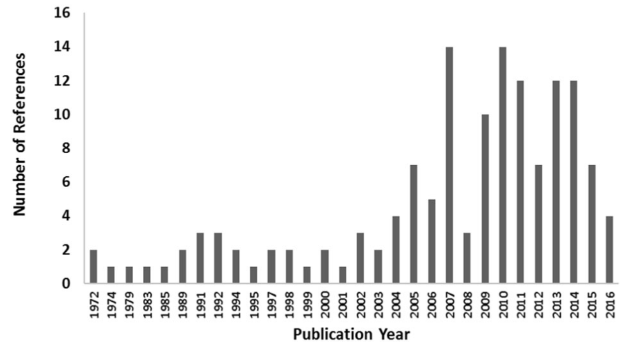

Fig. 2 Number of relevant references by publication year (EPRI 2018)

multiple sites. The large data sets were compiled from different state or regional resource agency monitoring programs, and the likelihood that the same data provided the basis for the reported analyses in different reports (double counting) appears to be negligible.

Most of the data sources described in the literature appear to be derived from field sampling programs that were implemented to support the development of fish consumption advisories and compliance with water quality standards. Therefore, large, predatory, recreationally important species that are more likely to be important sources of mercury exposure were the focus of most of these reviewed studies. The most common species in the reviewed temporal trend studies were: walleye (*Sander vitreus*), northern pike (*Esox lucius*) and largemouth bass (*Micropterus salmoides*). Walleye was noted in 36 of the reviewed studies, the northern pike was noted in 29 studies, and the largemouth bass was noted in 21 studies. Upon further review of fish consumption advisory monitoring programs conducted by individual states in the U.S., we found a general lack of monitoring data appropriate for temporal trend analysis. Most state monitoring programs do not provide data that are publicly, or otherwise readily available. In fact, many of the state monitoring data that were available for public download for inclusion in the Seafood Mercury Database in 2012, were no longer accessible at the time of our search. Also, many programs lack consistent, long-term monitoring data (for example, measurements of mercury in the same species, from the same water body, for at least 5 years), or they do not include data on potentially confounding factors (e.g., fish body size, trophic position).

The direction and magnitude of the trends in fish tissue mercury concentrations are the key indicators of change from this review. The reported increasing, decreasing and trend reversals represented statistically significant changes in fish tissue mercury concentrations. The proportion of decreasing trends exceeded the proportion of increasing trends by a factor of two (Fig. 4). Early in this literature review we identified the opportunity to quantify the

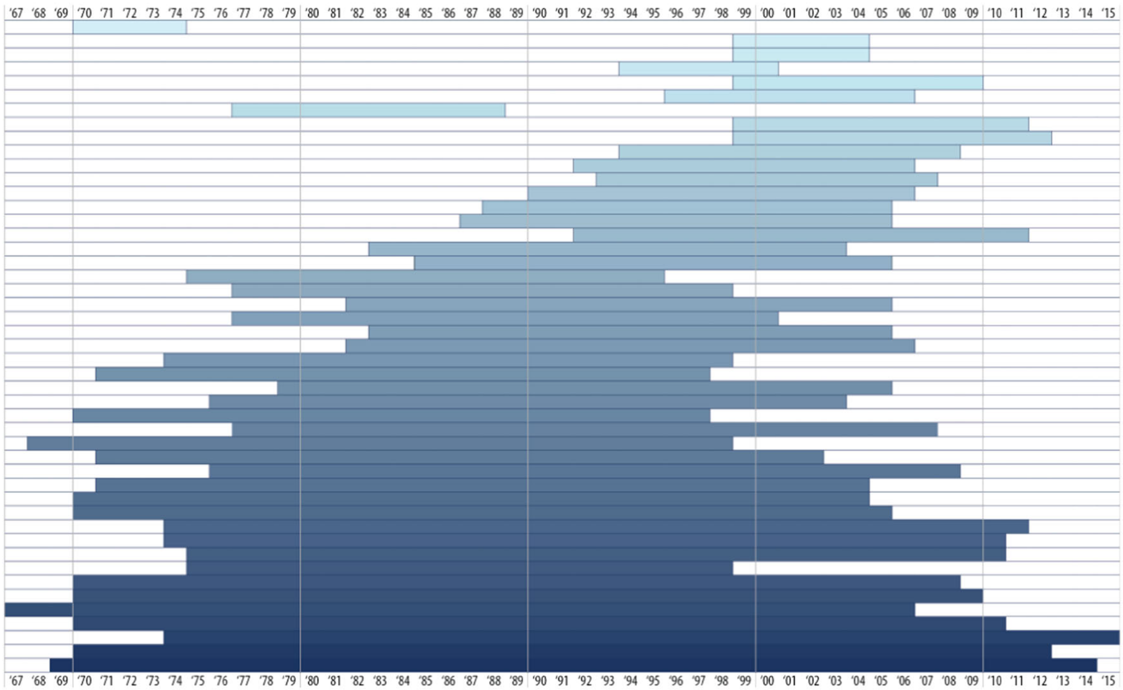

Fig. 3 North American freshwater fish studies ranked by duration of data record (EPRI 2018)

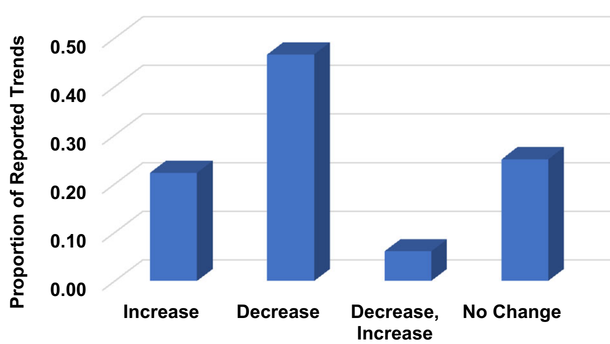

Fig. 4 Frequency of North American freshwater fish trends identified in reviewed literature (EPRI 2018)

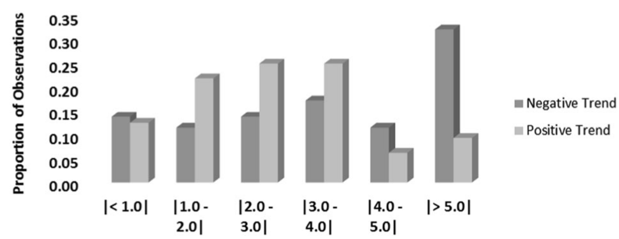

Fig. 5 Absolute values of annual percent change (APC) in the North American freshwater fish tissue concentrations by trend type (EPRI 2018)

differences in the magnitude of the observed trends in fish tissue mercury concentrations and to examine the relationship of these trends to the trends in atmospheric emissions of mercury and in mercury deposition concentrations. Based on the assumption of a constant rate of change over the measurement interval (linearity on the log scale), the annual percent change (APC) in fish tissue concentrations was used to compare the rates of changes in fish mercury levels among the available data sets.

Overall, we obtained 119 APC values from the 46 freshwater studies from the peer-reviewed literature. The mercury concentrations reported in these studies were based on standardized length measurements from individual species. The magnitude of the APC values associated with decreasing trends were much greater than the increasing trends. Figure 5 shows that a preponderance of higher APC values, e.g., values greater than 5% (absolute value), were associated with the decreasing trends. Overall, the average value for the reported or calculated APC was −4.2 for the negative trends (equivalent to a 34% reduction in fish tissue mercury concentration in 10 years). The average APC value for increasing trends in fish tissue mercury concentrations, reported or calculated from 32 data sets, was 3.0 (corresponding to a 25% increase in 10 years).

Several recent studies have shown that the observed declines in fish tissue mercury concentrations are either slowing down, levelling off, or reversing direction. Table 1 summarizes the results from the eight papers that reported changes in trend direction. The summary includes information on the geographic areas and species affected, the reporting periods, and the factors attributed to the observed change in trend direction. Generally, decreasing trends were reported during the period 1970–1995, and increasing

Table 1 Summary of results from studies showing a reversal in the trend in fish tissue mercury concentrations

Number	Reference	Location	Species	Period of record	All Species Affected	Hypothesized Factors
1	Monson (2009)	Minnesota Lakes	NP, WAE	1982–1992 1992–2006	Y	• Increased Hg deposition (global pool) • Climate change • Increased sulfate deposition
2	Monson et al. (2011)	Great Lakes Region	WAE, LMB	1982–1997 1997–2009	N	• Sample characteristics: age, gender, growth rates • Food web structural shifts, invasive species • Climate change
3	Ghandi et al. (2014)	Ontario, Canada	NP, WAE, LT	1970–1990 1995–2012	N	• Increased Hg deposition (global pool) • Climate change • Food web structural shifts, invasive species
4	Azim et al. (2011)	Great Lakes Region	WAE, YP, SMB, WB	1976–1995 1995–2008	N	• Food web structural shifts induced by invasive species • Climate change
5	Weis (2004)	Great Lakes Region	WAE, NP, YP, SMB, CC, WS	1971–1990 1990–1997	N	
6	Paller and Littrell (2007)	Southeastern U.S.	LMB, SF, CF	1971–2003 2003–2005	Y	• Local changes in hydrologic conditions, mobilization of atmospheric deposition of mercury
7	Sadraddini et al. (2011)	Great Lakes Region, Lake Erie	WAE, SMB, LT, NP, CC, RT, FWD, CS	1977–1997 1997–2007	N	• Food web structural shifts, invasive species • Increased Hg deposition (global pool)
8	Blukacz-Richards et al. (2017)	Great Lakes Region	WAE, LT, RS	1977–1997 1997–2007	N	• Climate change • Food web structural shifts, invasive species • Fluctuating water levels

trends were observed around the period 1995–2012. Monson (2009) reported that, before 1995, 64% of the 845 lakes sampled throughout Minnesota exhibited a decreasing trend in fish tissue mercury concentrations and 31% showed increasing trends. After 1995, mercury concentrations decreased in 35% of the lakes and increased in 60% of the lakes. The fish tissue mercury concentrations measured in the 845 lakes statewide over the period 1982–2006 are shown in Fig. 6. Monson (2009) noted that the observed upward trend in northern pike and walleye mercury concentrations beginning in the mid-1990s could be a response to the increased global emission of mercury between 1990 and 1995 (estimated by the author to be on the order of 1.2% year^{-1} in Minnesota) despite a decrease in regional emissions. Gandhi et al. (2014) also noted that the increased mercury emissions from Asia (which nearly doubled between 1990 and 2005) may have played a role in the observed increase in fish from the province of Ontario, Canada in recent years (1995–2012).

In all but two of the papers highlighted in Table 1, the increasing trends were not detected in all species examined in the study. For example, in the study of mercury accumulation in nine species in Lake Erie (Table 1, Study 7, Sadraddini et al. 2011) only three of the species (walleye, smallmouth bass and rainbow trout) exhibited statistically

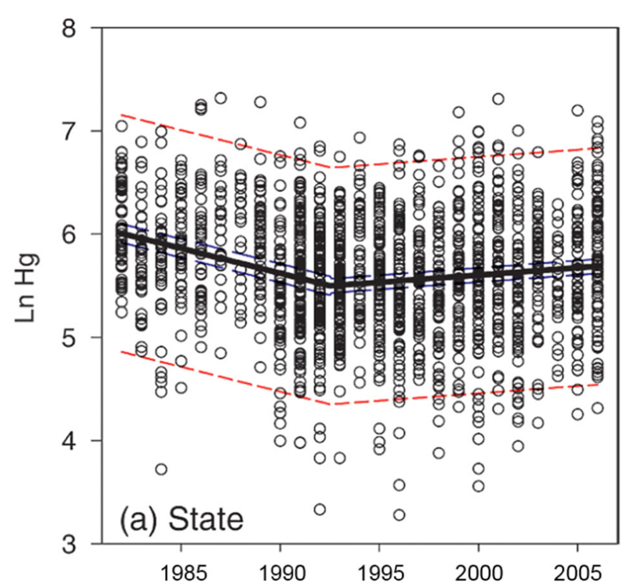

Fig. 6 Regression of natural log-transformed predator fish mercury concentrations from 1982 to 2006, showing a breakpoint in the trend in 1992 (Monson 2009)

significant increasing trends in their mercury levels after the mid- or late 1990s. In addition to increased atmospheric deposition, the other factors identified to potentially explain the observed mercury increases in the affected species were a systematic shift in the energy trophodynamics associated with the introduction of non-native species creating a new or expanding role for sediments as a net contaminant source. Azim et al. (2011) and others have noted the potential importance of the *Dreissenid* mussel and round goby invasions of Lake Erie and other Great Lakes that have created a new trophic pathway from sediment-bound mercury to benthic food webs, increasing mercury transfer to top predators.

The potential effect of climate change on the changing trends in fish tissue concentrations was noted by almost all the authors. Climate change factors such as increases in temperature (resulting in metabolic changes, higher rates of mercury methylation, and higher uptake rates of methylmercury), increased rainfall intensity and runoff (hydrologic export of organic matter carrying Hg^{II} from watersheds to surface water) and water level fluctuations that alter both the methylation of mercury or the mobilization of methylated mercury were suggested.

Freshwater fish tissue trends and mercury emissions

Mercury emissions in North America have decreased significantly over the period of record for the fish tissue samples reviewed in this study. For example, Fig. 1 shows that anthropogenic mercury emissions in the U.S. have declined from 223 Mg in 1990 to 92 Mg in 2005 to 51 Mg in 2011 (Weiss-Penzias et al. 2016; US EPA 2015). But during the period between 1990 and 1995 global anthropogenic emissions increased an estimated 19% (Pacyna et al. 2006). As noted above, Monson (2009) postulated that the observed increased trend in mercury concentrations in fish tissue from Minnesota lakes could be in response to the increased global emission of mercury between 1990 and 1995, despite a decrease in regional and North American mercury emissions.

Weiss-Penzias et al. (2016) also reported on trends in mercury wet deposition at 81 measurement sites of the MDN. They reported that between 1997 and 2013 there were significant negative trends in mercury wet deposition at 10 of the 19 sites (53%) and no positive trends. However, they also reported that between 2008 and 2013 with the addition of sites and greater geographic coverage, 5 (6%) of the sites exhibited negative trends and 24 (30%) of the sites exhibited positive trends. They also reported that, more recently, regional data from the Upper Midwest (UW) region of the U.S. displayed significant positive trends in mercury deposition. They hypothesized that these recent observations in the UW are consistent with a contribution from long-range transported mercury emissions.

As part of this study, we extended the Weiss-Penzias et al. (2016) analysis of the regional trends in mercury deposition with the most recent data from the UW region.

Table 2 Number of mercury deposition network sites in upper midwest region exhibiting positive and negative trends

	1996–2008	2008–2017
Number of Sites	7	10
Statistically Significant Negative Trend	2	0
Non-significant Negative Trend	4	1
Non-significant Positive Trend	1	5
Statistically Significant Positive Trend	0	4

The results for the period 2008–2017, summarized in Table 2, show that the reported increase in the occurrence of positive trends in mercury deposition among the measurement sites during the period 2008–2013 in the UW region are continuing. Statistically significant positive trends in mercury deposition over the period 2008–2017 were identified in 4 of the 10 regional sites, and none of the sites exhibited statistically significant negative trends.

Both fish tissue mercury concentration and atmospheric mercury deposition data included in this review have shown increasing trends, but the data do not indicate the existence of a linkage between the timing of the observed increases. The increasing trends in fish tissue mercury concentrations, reported in Table 1, were observed in the mid-1990s, but the increase in mercury deposition or reversal of the declining trend in mercury deposition in the UW region, where most of the studies showing a reversal in the trend in fish tissue mercury concentrations were reported, appears to occur around 2008 (Table 2). Not only did the observed increases in fish tissue concentrations occur earlier than the measured increases in mercury deposition, the data set for the fish tissue mercury concentrations only extends to about the period that the trend reversal in deposition was observed (circa 2008). Although several studies have shown a rapid response in biota mercury concentrations to decreases (Hutcheson et al. 2014) or increases in atmospheric mercury inputs (Paterson et al. 2006; Harris et al. 2007), the response of mercury tissue concentrations in large predatory fish species can exhibit a delay from increased atmospheric loading. Results from the METAALICUS project (Harris et al. 2007; Hrenchuck 2018; Blanchfield 2019) showed that mercury added to the lake surface was measured in young-of-the-year yellow perch within two months. For the larger predatory species, it took 3 years for the isotopic mercury added directly to the lake surface to account for 10% of the overall total mercury concentration in northern pike and lake whitefish. The METAALICUS study also showed that very little of the mercury added to the terrestrial system made it to the lake over the first eight years of monitoring. Modeling results from the study of the response of predatory fish tissue mercury in a freshwater lake to changes in mercury deposition (Vijayaraghavan et al. 2014) suggest that it would take 3–8 years for a change in deposition to begin to be reflected in fish tissue mercury concentrations.

Assessment of trends in marine fish

A primary motivation for monitoring fish tissue mercury concentrations in marine species is the importance of estuarine and marine fish consumption as a pathway of mercury exposure. The primary source of mercury exposure in the human diet in North America is from the commercial fish market (Sunderland 2006), and estuarine and marine species comprise >90% of the fish in the commercial market (Carrington et al. 2004). Additionally, methylmercury concentrations in commonly consumed marine fish species exceed the U.S. EPA human health criterion of 0.3 ppm in most marine systems studied (Karimi et al. 2012; Chen et al. 2012).

Of the 39 marine studies that were identified in the literature search, 25 references dealing with particular species of fish included information on the study period and species studied as well as an evaluation of trend direction. The average time between the first and final observation in the trend studies was 30 years, and the duration of the studies ranged from 6 years to 93 years. All but four of these studies extend over a period of at least 10 years.

Overall, the reviewed studies reported on fish tissue mercury measurements in more than 14,000 individual or composite samples, and one study (Davis et al. 2016) accounted for 6,539 measurements, or almost half of the total reported marine fish tissue mercury measurements. By comparison, over one million fish samples were analyzed in the freshwater literature review discussed above. The 25 studies that are the focus of our more detailed analysis on the literature included 19 species.

The reported trends in the tissue concentrations of mercury in marine fish were dominated by two categories of trends, either "decreasing" or "no discernible". A decreasing trend was reported in 11 fish species, and a summary of the results from these published studies is presented in Table 3. Most of these studies (Studies 5–11) report on findings from the nearshore or continental-shelf environment. Moreover, Studies 5–9 report on the observed decreasing trends in four fish species measured in the eastern region of the Baltic Sea, where the highest mercury concentrations in fish were measured from the fishing grounds in close proximity to riverine inputs (Polak-Juszczak 2013). The decreasing trends in fish tissue mercury concentrations were attributed to reductions in both river runoff to the Baltic and in atmospheric deposition (Polak-Juszczak 2013).

The identification of declines of mercury in bluefish from the mid-Atlantic coast of North America (Study 1 in Table 3, Cross et al. 2015) is dependent upon data from multiple sources, and the earliest reported elevated mercury concentrations in fish tissue from 1972 provides an inconsistent initial high-value point that potentially affects the overall trend-analysis results. In another recent analysis of mercury and other toxic pollutants in marine fish based on data compiled from multiple sources (Study 2 in Table 3) Bonito et al. (2016) reported an overall statistically significant negative trend in fish tissue concentrations measured in 841 fish samples collected globally. However, the coefficient of determination in the regression analysis (r^2) was 0.03, and a statistically significant regional trend was detected in only one of the five geographic areas tested separately (Atlantic Ocean). Furthermore, the Bonito et al. (2016) study presents average values for a wide variety of fish species (several hundred species), and no consideration is given for species, size or age of the fish in their compilation—all of which are well-known to be factors significantly influencing mercury concentrations in fish.

In Study 3 (Table 3) there is a greater emphasis on spatial and species differences in fish tissue mercury concentrations from the near-coastal U.S. waters of the Atlantic and Gulf of Mexico than on an analysis of temporal trends (Adams and McMichael 2007). The authors did report that although the temporal, spatial and fish-size distribution of the target species did not allow for rigorous time-series analyses, they used two collection periods (pre-1996 and post-1996) for the analysis of temporal changes in mercury levels of Spanish mackerel. This analysis indicated the existence of a statistically significant decrease in fish tissue mercury concentrations between the measurement periods, but the authors also reported that mercury levels in the tissue samples collected between 1990 and 2002 were higher than the values from fish collected during the 1970s.

Study 4 in Table 3 (Lee et al. 2016) is based on measurements of mercury in samples from 1292 samples of wild-caught bluefin tuna in the North Atlantic over an 8-year period from the 1990s to early 2000s in which atmospheric loading of mercury from North America declined. The results show that fish tissue mercury concentrations in age-normalized fish declined 19%. This decrease in fish-tissue concentrations parallels the reduction in anthropogenic emission rates in North America. The authors note that this report provides "the first evidence to suggest that emission reduction efforts have resulted in lower mercury concentrations in large, long-lived fish". Importantly, the data in Lee et al. suggest that Atlantic bluefin tuna responded rapidly to changes in mercury emissions from North America, which in turn resulted in declines in mercury concentrations in air above the North Atlantic and Atlantic surface waters. These results were consistent with the trend observed over a longer period in bluefish caught in mid-Atlantic waters (Cross et al. 2015).

Table 3 Summary of studies that showed decreasing trends

Study number	Reference	Location	Species	Period of record	Key information
1	Cross et al. (2015)	Mid-Atlantic Coast of U.S. Atlantic	Bluefish, *Pomatomus saltatrix*	1972–2011	• 43% decline in [Hg] between 1972 and 2011 • Corresponds to decreases in elemental Hg in North Atlantic Ocean
2	Bonito et al. (2016)	Global, Atlantic Ocean Region	Multiple species, four trophic guilds	1969–2012	• Large data set, n = 841 • Significant trend, small r^2 value
3	Adams and McMichael (2007)	Atlantic Ocean, Gulf of Mexico	Spanish mackerel *S. maculatus*	1990–2002	• Comparison of pre- and post-1996 data indicate that Hg concentrations have declined between 1990 and 2002 • Hg levels higher than in 1970s
4	Lee et. al. (2016)	North Atlantic	Bluefin tuna, *Thunnus thynnus*	2004–2012	• Hg concentrations declined 19% from 1990s to early 2000s (2% per year) • Large data set, n = 1292
5	Polak-Juszczak (2010)	Atlantic, Baltic Sea	Flounder, *Platichthys flesus*	1996–2003	• Hg sources include both coastal runoff and atmospheric deposition
6	Polak-Juszczak (2013)	Atlantic, Baltic Sea	Cod, *Gadus morhua*	1994–2010	• Hg sources include coastal riverine input, atmospheric deposition and legacy sediment concentrations • 57% reduction in muscle tissue Hg concentrations
7	Polak-Juszczak (2009)	Atlantic, Baltic Sea	Herring, *Clupea harengus*	1994–2003	• Hg sources include coastal riverine input, atmospheric deposition and legacy sediment concentrations
8	Polak-Juszczak (2009)	Atlantic, Baltic Sea	Sprat, *Strattus spratus*	1994–2003	• Hg sources include coastal riverine input, atmospheric deposition and legacy sediment concentrations
9	Polak-Juszczak (2009)	Atlantic, Baltic Sea	Cod, *Gadus morhua*	1994–2003	• Hg sources include coastal riverine input, atmospheric deposition and legacy sediment concentrations
10	Guns et al. (1992)	Atlantic, North Sea	Flounder, *Platichthys flesus*	1971–1990	• Fish samples from Belgian continental shelf waters
11	Guns et al. (1992)	Atlantic, North Sea	Plaice, *Pleuronectes platessa*	1971–1990	• Fish samples from Belgian continental shelf waters

Table 4 Summary of studies that showed increasing trends

Study number	Reference	Location	Species	Period of record	Key information
1	Drevnick et al. (2015)	North Pacific Ocean (NPO), Hawaii	Yellowfin tuna, *Thunnus albacares*	1971–2008	• Rate of increase since 1998: 3.8%/year, in agreement with changes in Hg cycling in NPO
2	Drevnick and Brooks (2017)	North Pacific Ocean (NPO), Hawaii	Yellowfin tuna, *Thunnus albacares*	1971–2008	• Reanalysis of data from 2015 study; additional data included in analysis • Rate of increase 1998–2008: 5.5 %/year
3	Drevnick and Brooks (2017)	North Pacific Ocean (NPO), Hawaii	Bigeye tuna, *Thunnus obesus*	1971–2008	• Rate of increase 1998–2008: 3.9 %/year
4	Burger and Gochfeld (2004)	Global	Skipjack tuna, *Katsuwonus pelamis*	1991–2003	• Values in canned tuna for 1998–2003 significantly higher than 1991 measurements

Table 4 summarizes the results from the published studies that reported increasing trends in fish tissue mercury concentrations. Studies 1–3 report on the analysis of mercury in yellowfin and bigeye tuna caught in the North Pacific. These reports are based on the compilation of data from six different data sources (Drevnick and Brooks 2017). The authors noted that the data unequivocally show an increase in fish tissue mercury concentrations with time, but they also note that the limited sampling and variability in mercury concentrations introduce uncertainty and affect the overall understanding of fish tissue trends. The fourth study identified in Table 4 is based on measurements made of canned tuna (Burger and Gochfeld 2004). One out of three different types of tuna exhibited an increasing trend.

Both the limited number of trend studies and the details of the studies that do identify a trend presented in Tables 3 and 4 indicate a general paucity of information on marine fish tissue concentrations. Most of the identified published results are combined with data from previous reports. The results from a recent attempt to compile data on fish tissue mercury concentrations from coastal waters of California and western North America is instructive (Davis et al. 2016). The study developed a data set that included a total of 6,539 individual records for fish mercury concentrations sampled between 1985 and 2014. Most of the locations sampled had a species with an average concentration above 0.3 μg/g wet weight. But there were limited data on long-term trends. Davis et al. (2016) noted the lack of availability of data in state and federal databases. They also called for systematic and consistent monitoring as well as increased access to collected data to monitor trends in fish tissue mercury concentrations in a changing environment.

Summary

Significant overall decreasing trends in fish tissue mercury concentrations in North American lakes were reported over the period 1972–2016. More recently, a leveling off in the rate of mercury tissue concentration decreases or increases in tissue concentrations have been reported. Increased emissions of mercury from the global pool beginning between 1990 and 1995, despite a decrease in regional emissions, have been advanced as an explanation for the observed changes in fish tissue trends. In addition to increased atmospheric deposition, the other factors identified to explain the observed mercury increases in the affected fish species include a systematic shift in the food-web structure with the introduction of non-native species, creating a new or expanding role for sediments as a net contaminant source. The influences of climate change—such as increases in temperature (resulting in metabolic changes and higher uptake rates of methylmercury), increased rainfall intensity and runoff (hydrologic export of organic matter carrying Hg^{II} from watersheds to surface water) and water level fluctuations that alter both the methylation of mercury or the mobilization of methylated mercury—have also been identified as contributing factors.

Opportunities exist to design more efficient, informative long-term fish monitoring programs to detect trends and to relate fish tissue mercury measurements to potential influencing factors. New or revised monitoring efforts should address several factors. First, monitoring data should be made publicly available whenever possible to facilitate trend analysis, such as the comparison of trends within and among regions. Second, the design of future monitoring program should consider the magnitude of the changes expected and how long it will take to detect a statistically significant change with a specified level of confidence. Our review of the statistical techniques used in the analysis of the freshwater fish tissue mercury trends showed the effective use of both traditional linear regression and innovative Bayesian inference techniques. Third, monitoring efforts must include the measurement of important, potentially confounding factors (e.g., fish body size, fish trophic level, potential mercury sources such as aqueous or atmospheric mercury data) that can change over time, and are known to be related to fish mercury content. Ideally, modern temporal trend analysis should also include factors that allow us to understand the role of changes in climate in influencing fish mercury concentrations over time.

The primary source of mercury exposure in the human diet in North America is from the commercial market which is dominated (>90%) by marine species, but by comparison very little information is available on mercury trends in marine fisheries. Most of the data used in the published marine trend studies are cobbled together from previous reports. The data collection efforts are intermittent, and the spatial and fish-size distribution of the target species vary widely. As a result, convincing evidence for the existence of fish tissue mercury trends in marine fish is generally lacking. However, there is some evidence from sampling of large, long-lived, commercially-important fish showing both lower mercury concentrations in the North Atlantic in response to reduced anthropogenic mercury emission rates in North America and increases in fish tissue mercury concentrations with time in the North Pacific in response to increased mercury loading. Complicating the relationship between mercury concentrations in fish and mercury loadings to ecosystems are the ecological factors that may influence fish tissue concentrations. Thus, changes in the mercury concentration in key dietary components that result from changing fishing pressures, disease in natural populations, or changes in physical and chemical factors that may alter the mercury levels in available prey populations, may affect mercury levels in predatory fish. Therefore, less direct relationships between mercury loadings and fish concentrations may sometimes occur.

Additional measurement of mercury levels in marine fish, standardization in approaches and interpretations of trends and monitoring and modeling studies that address the causes of fish mercury trends are all necessary to improve our understanding of fish mercury and human exposure. However, the findings from this study suggest that efforts to reduce mercury emissions can have rapid consequences in comparable reductions in mercury concentrations in marine fish. Thus, it is possible that human remedial actions to reduce mercury loadings into aquatic ecosystems can have significant effects on seafood mercury concentrations. It seems imperative to establish a baseline of methylmercury in key marine species to evaluate whether changes are detectable in subsequent years. The time is right to institute such monitoring efforts as the Minamata Treaty (entered into effect in 2017) should result in large changes in mercury emissions, particularly from industrialized countries.

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Compliance with ethical standards

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